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Alpha radioactivities from nuclides in the atomic number region 80–83

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ABSTRACT

By bombardments of targets of wolfram, rhenium, osmium and iridium with carbon ions in the Stockholm 225-cm cyclotron, a great number of light nuclides, mainly belonging to the elements mercury, thallium, lead and bismuth, has been produced. Possible alpha emitters have been searched for and the results obtained hitherto are preliminarily reported in this paper.

Introduction

Up to 1947 very little was known about alpha-radioactive nuclides with mass numbers less than 210 [1, 2]. Some unpublished data were, however, available, mainly in the United States. In the following few years several investigations were reported.

Two different atomic number regions became of interest in this respect [3]. One of them was the rare earth region, with light nuclides of the elements from cerium to holmium ($Z = 58–67$). The other one, which will be further discussed in this paper, was the region just below the heavy elements among which alpha activities were well known from studies of the four, now classical decay series.

Some new light polonium isotopes were studied by Templeton, Howland and Perlman [4]. The results were published in 1947. The recent developments in the field of neutron-deficient polonium isotopes have been discussed in earlier papers from this institute [5–8].

Naturally occurring platinum was first reported by Porschen and Riezler to have some feebly alpha-radioactive isotopes [9, 10].

In 1949, Thompson, Ghiorso, Rasmussen and Seaborg preliminarily communicated results obtained in the bombardment of a gold target with 190 MeV deutrons in the Berkeley 184-inch cyclotron [11]. Two short-lived alpha activities had been found. Later, this group gave a detailed account of these experiments [12]. One of the activities was assigned to a light gold isotope, the other to a mercury isotope. The last-mentioned one had a half-life of only 0.7 minutes and an alpha-particle energy of 5.60 MeV. This activity has recently been confirmed and some other mercury alpha activities have been discovered by Karras, Andersson and Nurmia in Uppsala from proton bombardments of gold in the synchrocyclotron of the Gustaf Werner Institute for Nuclear Chemistry [13]. They have all relatively short half-lives and are assigned to mass numbers in the region below 191.

As far as the present author is aware, no alpha radioactivities have hitherto been reported for nuclides of the elements thallium and lead.

In 1948, Templeton and Perlman [14] briefly discussed some new artificial alpha-radioactive bismuth isotopes obtained from bombardments of lead with 100–200 MeV deutrons in Berkeley. A lack of precision in the alpha-particle pulse analysis was, as stressed by the authors, caused by the high background of electrons and the difficulty in preparing extremely thin samples. These problems have also been inherent in the investigations of other groups [5].

A few years later, Neumann and Perlman [15] published an extensive study of the decay properties of neutron-deficient bismuth isotopes in the mass number region 198–203. Mass number assignments were made, based on the study of genetic relationships with earlier known lead and thallium isotopes, in this case formed by electron capture decay of purified bismuth fractions. The degree of certainty in the mass number assignments was still not considered to be very high, as is seen from the classification code letters in the 1958 Berkeley table of nuclides [16].

From the beta-spectroscopic point of view, this neutron-deficient region has been extensively studied by, for instance, Andersson and co-workers in Uppsala [17]. The Chacketts have recently studied light thallium isotopes produced by bombardments of wolfram with nitrogen ions [18].

At the Nobel Institute of Physics, neutron-deficient nuclides in the element region mercury-bismuth ($Z = 80$–83), have, during the last few years, been produced by bombardments of different targets with carbon ions and, in a few cases, also with neon ions. The present paper is a preliminary report of data obtained by the heavy ion research group of this institute from studies of the alpha-radioactive properties of the type of nuclides mentioned.

Experimental techniques

Most bombardments in the present investigation were made by use of the internal carbon ion beam in the 225-cm cyclotron of the Nobel Institute of Physics [19, 20].

The accelerated and utilized ion was $^{12}C^{4+}$, produced from carbon dioxide. Neon ions have also been used in a few cases, but the yields of the products obtained were generally too low for the investigations under consideration here. The bombardments were made at radii between 85 and 90 cm. The energies and intensities of the beams obtainable in the cyclotron accelerations have been discussed in earlier papers [19, 5, 7]. A radius often used for the carbon ion bombardments was 88 cm. In this case most of the $^{12}C^{4+}$ ions had energies between 110 and 125 MeV. The beams reaching the target holder with these energies were in the range of 10^{-2}–10^{-1} μA. The bombardments lasted from a few minutes to about two hours.

In this investigation most of the bombardments were made by use of the recoil collection technique. The procedure has earlier been described in detail [5, 7, 19]. In many of the runs, however, a modified target holder has been used, to which larger, rectangular targets and catcher foils could be applied. Compared with what has earlier been possible, a greater part of the beam has in this way been utilized in the bombardments.

Targets of tantalum, wolfram, rhenium, osmium and iridium have been used in this study. Several methods have been applied in the preparations of these targets. These methods, as well as the different chemical separation and preparation procedures which we have employed in this element region, will be described in more detailed communications. The sample preparation technique used was in many cases of fundamental importance for the results obtained.

The alpha-radioactivity measurements were made with a grid ion chamber and a multichannel pulse-height analyzer. The instruments have been described by Åström [21]. An automatic recording system used in most of the present work made the registration of every alpha count possible with respect to energy and time of occurrence. This system has also been described elsewhere [22].

In a few cases, the electromagnetic isotope separator of this institute [23, 24] has been used in attempts to make mass number assignments of the activities studied in the present investigation. Hitherto these efforts have been unsuccessful mainly because of the relatively low yields of the reaction products after the bombardments. In some cases, however, there is good hope of obtaining more favourable yields in the near future and thus of making final mass number assignments possible. Until then, the detailed descriptions of some of the experiments reported here will probably be deferred.

Survey of the results

The bombardments of tantalum targets with carbon ions have hitherto given no alpha-radioactive products. Such have, however, been found from the various nuclear reactions surveyed below. In the reaction formulae, x is the number of neutrons, n, evaporated from the compound nucleus in question.

Products from W(C,xn)Hg

The vaporization method [12] used for the sample preparation after the bombardment showed that some alpha activities produced were due to mercury isotopes. Four not fully resolved intensity peaks have been found in the alpha spectrum in the particle-energy region 5.10–5.35 MeV. Fine structure possibly occurs in some cases. The approximate half-lives of the decaying mercury isotopes suggest, according to the electron-capture decay data given by earlier investigators [16], that they have mass numbers in the region 191–195. Such mass numbers are also expected from the nuclear reaction conditions.

In the mass number region just below, Karras, Andersson and Nurmia [13] have recently found an interesting trend in the alpha-decay energies as a function of mass number for mercury isotopes. Our results seem to continue this trend.

Products from Re(C,xn)Tl

Two alpha activities were found after these bombardments. One was too short-lived ($t_{\frac{1}{2}} \sim 3$ min) and formed in too low a yield to make alpha-energy measurements possible in the ion chamber after the necessary preparation. The activity was found directly from the target foil by use of a scintillation detector [7]. For this reason its chemical identity cannot be stated, but possibly it is due to ^{190}Tl or ^{191}Tl.

The other alpha activity found from the above-mentioned nuclear reaction had a half-life of about half an hour and an alpha-particle energy of about 5.80 MeV. This activity is probably due to ^{193}Tl or ^{194}Tl. The mass numbers suggested are in agreement with what could be expected from the nuclear reaction data.

Products from Os(C,xn)Pb

In this case rather more unexpected results were obtained. From the products of the osmium bombardments two rapidly decaying alpha activities were found. One of them had an alpha-particle energy of about 6.7 MeV and a half-life of approximately two minutes. The other one had an energy of 6.4 MeV and a half-life of about four minutes. Because of the short lifetimes chemical separations could as yet not be carried through. A comparison of the alpha-particle energy spectra in this case with those obtained in the other studies in this series give no evidence for the suspicion that, for instance, impurities in the osmium targets could have given product nuclides of bismuth or polonium. Still less reasonable is the idea that the alpha activities under consideration could be due to elements with lower atomic numbers than lead since, for instance, the mass numbers obtainable from the nuclear reactions are here too high to fit the available decay data in a lower region. As the α/EC-branching ratios are not known in the present cases, no partial alpha-decay half-lives can be discussed here.

At particle energies of about 6.0 and 5.8 MeV two further, earlier unreported, weak alpha activities with longer half-lives appeared. They should probably be assigned to some lead isotopes in the mass number region 196–199.

Products from Ir(C,xn)Bi

The alpha activities of light bismuth isotopes found by Neumann and Perlman [15] have mainly been confirmed at this institute [5], even if the particle energy values will have to be slightly modified. Also, no definite mass assignments have yet been made by us.

Among alpha activities in bismuth, which can be ascribed to isotopes with mass numbers less than 198, there are some new data to report. Thus, we have found intensity maxima in our alpha-particle energy spectra at approximately 6.8, 6.7, 6.45, 6.35, 6.16 and 6.08 MeV. The objection could be raised that some of these could be due to lead isotopes mainly formed by electron-capture decay of the bismuth isotopes during the time of the experiment. This could, for instance, seem to be reasonable for the alpha activities having the energies 6.7, 6.45 and 6.35 MeV and the half-lives very close to those reported above for the short-lived lead activities, if the mother nuclides are also assumed to be very short-lived.

Contrary to this suggestion, it can be pointed out that, if the lead isotopes, discussed in the previous section, have mass numbers below 194, which is the mass number of the most neutron-deficient lead isotope hitherto reported, with a half-life of about 11 minutes [25], then, these isotopes cannot have been produced under the energy conditions prevailing in the iridium bombardments as the lightest stable iridium isotope in the target is ^{191}Ir.

The bismuth istotopes formed must, for the reasons indicated above, all have mass numbers greater than 193. The occurrence of six different alpha-energy values in such a narrow mass number region suggests the occurrence of fine structure in the alpha-particle energy spectrum. The half-lives found for some of the intensity peaks are close to three minutes.

It should be remembered that this is a preliminary report in which the experimental data given in a few cases, even if they are based on several bombardments, are not to be considered as definite. In the bismuth samples the counting difficulties due to the overwhelming amount of electron-capture decay should call for cautiousness in the statements.

Finally, it should be mentioned that more detailed alpha-decay data are available for bismuth in the region 5.0–6.0 MeV and will be published by us after further efforts to obtain samples useful for mass assignments have been made.

Conclusion

The task of the presented study, to search for alpha activities in the element number region 80–83, has undoubtedly given some interesting results in itself. Certainly, several more new alpha-emitters will also be found in the studied region in future experiments. However, this work was started as a complement to some other investigations. In several papers from this institute, alpha-decay properties of polonium and astatine nuclides have been reported [5–8, 22]. Even if the chemical separations and the isotope separations carried out in these investigations were generally considered to give reliable results, it was thought that a map drawn of the alpha activities in the mass region 190–200 of the elements with atomic numbers 80–83 would add to the confidence in the data reported. Possible contaminations occurring in the targets used and in the samples of the products studied, for instance from daughter nuclides, could in this way be more easily revealed.

The experiments have shown that there is a neutron-deficient region, in which the alpha-decay properties and the mass numbers for nuclides of polonium, bismuth and lead are very similar, and in which the possibilities for accurate determinations are limited because of the short lifetimes involved.

In the alpha-particle energy range of 6.1–6.8 MeV there are, thus, several nuclides of the enumerated elements which have half-lives from about five minutes down to fractions of a minute. In some of these cases only reasonable suggestions of the identities of the products can be made from comparisons with what has been learned in earlier experiments and from the nuclear reaction data under consideration. The possibilities for more reliable statements are increased if, as in the present case, a series of similar irradiations has been made with targets of neighbouring atomic numbers.

In the range of lifetimes discussed, the time available for definite chemical identification is often too short and the use of an alpha-daughter recoil milking device of the type suggested by Ghiorso [26] will probably be of limited value because of the interference of the electron-capture decay daughters in this region.

Studies of the growing in of alpha-radioactive daughter products have been made in some cases, where the mother nuclide has first been isolated [22]. In the present study, data for such daughter products have been obtained from the mercury samples.

It can finally be mentioned that the alpha activities reported in the present paper cannot have been responsible for contaminating radioactivities in our earlier investigations of the higher atomic number region.

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